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LA-UR -88-1991

CONF-880631--45

Received by OSTI

4 1988

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LA-UR--88-1991

DE88 014435

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SUBMITTED TO 29th Annual Meeting of the Institute of Nuclear Materials Management, Las Vegas, June 26-29, 1988

MASTER

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THE USE OF PROCESS INFORMATION FOR VERIFICATION OF INVENTORY IN SOLVENT EXTRACTION CONTACTORS IN NEAR-REAL-TIME ACCOUNTING FOR REPROCESSING PLANTS

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ABSTRACT

Near-real-time accounting is being studied as a technique for improving the timeliness of accounting in nuclear fuel reprocessing plants. A major criticism of near-real-time accounting is perceived disclosure of proprietary data for IAEA verification, particularly in verifying the inventory of solvent extraction contactors. This study indicates that the contribution of uncertainties in estimating the inventory of pulsed columns or mixer settlers may be insignificant compared to uncertainties in measured throughput and measurable inventory for most reprocessing plants, and verification may not be a serious problem. Verification can become a problem for plants with low throughput and low inventory in tanks if contactor inventory variations or uncertainties are greater than ~25%. Each plant must be evaluated with respect to its specific inventory and throughput characteristics.

INTRODUCTION

Near-real-time accounting (30-day or less balance closure periods) has been proposed as a technique for improving the timeliness of accounting in reprocessing plants. For both conventional and near-real-time accounting, the operator must measure all transfers into the facility and out of the facility, and the inspector must verify these measurements. For conventional accounting the in-process inventory is measured once per year by draining out the facility.

For near-real-time accounting one must verify the transfers as for conventional accounting. The major difference between near-real-time accounting and conventional accounting is that the in-process inventory must be measured or estimated on a more timely basis. The Agency now considers accounting periods of approximately one month; therefore, all nuclear material residing in process tanks, solvent extraction contactors, and concentrators must be estimated or measured by the operator, and these measurements must be verified by the inspector, at least on a monthly basis.

*Work supported by the U.S. Department of Energy, Office of Safeguards and Security.

One major problem in applying near-real-time accounting for reprocessing facilities arises from the requirement for the inspector to verify the material in the process. A particular problem arises in estimating inventory in the solvent extraction contactors whether they be mixer settlers or pulsed columns.

Two techniques have been proposed for determining the amount of nuclear material in the contactors. One method relies on estimation of the contactor inventory from process operating conditions. Considerable work in developing theoretical estimation models has been performed, for example, by Beyerlein and Geldard at Clemson University,[1] by Cobb at Los Alamos,[2] by Burkhart at Iowa State,[3] and by Japanese and IAEA workers.[4] Estimation of solvent contactor inventory from process data is being used at the fast breeder fuels reprocessing plant at Dounreay and is the proposed method for the BNFL light-water reactor (LWR) processing plant, THORP, under construction at Sellafield. The problem arises in that many operators consider much of the information that would be required for these theoretical models to be proprietary, and they are reluctant to give the information to the IAEA.

The second method for determining the inventory in contactors is by direct measurement. This was investigated by Ehinger at the Barnwell Reprocessing Plant in the late 70s and early 80s.[5] In this technique density probes are inserted into the column. From the known flow rates of the aqueous and organic streams, one can calculate the density attributable to the liquid in the columns. The remainder of the density is attributable to the nitric acid and heavy metal concentration. By filtering the noise from the pulsing action on the columns, one can get a reasonable measure of the heavy metal content in the columns. This method also may suffer from reluctance on the part of facility operators to make the density probes accessible to the IAEA inspector and from a perceived inability on the part of the inspector to independently verify the concentration derived from the density probes.

THE VERIFICATION PROBLEM

Regardless of how the plant operator determines the contactor inventory, the inspector has the problem of verifying the operator's data.

Hamlin[6] proposed that in-process holdup be used as a safeguards measure. He suggested that any process is designed to operate within a limited holdup range, and higher or lower holdup, as indicated by comparing process input and output, is indicative of abnormal operation. Thus, potential diversion can only be accomplished within this normal range; any larger diversion would be detectable by statistical techniques. He states, "If that part of the in-process inventory that is only measurable by input/output analysis has an upper operating limit by less than a significant safeguards quantity of the material in question, the IAEA's criteria for both quantity and timeliness can be met by a combination of input/output analysis to determine in-process holdup during the campaign, together with a material balance over the campaign." He discusses procedures to reduce in-process inventory during materials balance closure to attempt to reduce the holdup range.

Delange[7] is applying such an approach, referred to as cumulative flux or running book inventory, at the reprocessing plant at LaHague. However, the in-process inventory and the normal allowed variation in in-process inventory are large and greatly exceed the proposed significant safeguards quantity of 8 kg.

Walford et al.[8] attempted to define the expected normal operating variation in the in-process inventory of solvent extraction contactors. The pulsed-column contactor operation was computer-simulated using the SEPHIS code[9] for a flow sheet reprocessing fast breeder fuel. For normal operation and reprocessing at a rate of 0.4 Te/day (~100 kg plutonium per day), the first cycle contactor inventory is 1 kg of plutonium and can increase to ~4 kg without serious process impact. However, a higher inventory results in significant plutonium loss to the waste stream. For LWR fuel, the first cycle contactor inventory of plutonium would be significantly less, probably ~50 g. For mixer-settlers, the inventory would be smaller than for columns. In any case, the contactor inventory is less than one significant quantity for a plutonium throughput of 250 kg/day.

We are proposing the use of process information for determining the amount of plutonium in the solvent extraction contactors in the plutonium purification cycle of the reprocessing facility.

The process flow sheet dictates an approximate concentration level of plutonium in each of the four contactors in the plutonium purification cycle. Each column will have some variability to this inventory, again dictated by potential variations in parameters such as organic and aqueous flow rates into each of the columns and plutonium concentration into the 2A column. The plutonium concentration into the 2A column will vary generally with the type of fuel being processed, that is, whether BWR or PWR fuel is being processed, and the burnup of the fuel.

Whether process design information can be applied to the solvent extraction contactors will depend on several parameters. The total uncertainty in measurements for near-real-time accounting will be a combination of errors associated with transfers through the process and errors associated with measurement or estimation of material in the process. For material in the process, if the amount of material in contactors is small compared to the amount of material in measurable items such as tanks, the errors in the tanks will tend to dominate, and errors for the contactors may become insignificant. Also, if the amount of material in the contactors is small compared to the throughput through the facility, errors associated with transfer measurements will tend to dominate, and errors associated with the contactor inventory will tend to be small, relatively.

The question of whether process design information for the extractors can be applied to the solvent extraction contactors then reduces to a question of the relative contribution of errors associated with the contactors, the remainder of the process tanks, and the transfer measurements. The characteristics of the materials balance equation must be examined on a plant-specific basis to determine the applicability of this approach.

CALCULATIONS

We have modeled the error contributions from measured throughputs (input and output), measurable inventory in process tanks, and unmeasurable inventory in solvent extraction contactors. The model covered the range of throughputs and inventories in existing commercial reprocessing plants and those plants expected to be in operation by the end of the century. The study indicates that for many cases the uncertainty in plutonium content of solvent extraction contactors is small compared to uncertainties in measured transfers and measurable inventory. It is suggested that in those cases contactor inventory can be inferred from process operators' data and need not be verified by measurement during process operation.

References 10 and 11 detail the methodology used to conduct the analyses described in this report. The assumptions made and the limitations imposed by these assumptions are described below.

The variance equations used for this analysis assume steady-state facility operation. The series of batch transfers for each stream (inputs and outputs) and for each process unit inventory measurement are assumed to have the same (constant) nuclear materials concentration measurement and volumetric measurement for any single stream or vessel. The effect of this assumption is to understate the total variance for each individual stream or vessel inventory. Occasions arise each as during plant startup or shutdown for cleanout, where the assumption of steady-state operation is not valid.

The assumption of steady-state operation yields less accurate answers for systems where the MBA contains few process units or where the length of the accounting period (number of batches processed) is short. In the systems that involve smaller MBAs, we have found that the non-steady-state model may show variances up to 50% greater than the steady-state model. The Safeguards Systems Group has found that the assumption of steady-state operation is an acceptable one for sensitivity studies on reprocessing plants.

The model assumes that measurements of batch concentration or volume are correlated for each vessel. This assumption can result in an overstatement of the overall variance associated with each vessel. However, the model assumes no correlation between measurements on separate streams or different vessels, which may lead to an understatement of the combined variance for all vessels because samples taken from a series of vessels may all be measured on a single or limited set of infrequently calibrated instruments. No recalibration is assumed to occur during an accounting period.

The model also assumes that the contactor inventories vary randomly and are not correlated. These limitations on the model should be considered when applying the results of this study. Each plant should be evaluated on the basis of its particular characteristics.

RESULTS

The total system (transfer and inventories) standard deviation for a 30-day accounting period was plotted as a function of contactor inventory (0–20 kg) and tank inventory (0–200 kg) for facilities of low throughput (5 kg/day, 100 kg/30-day accounting period) and high throughput (50 kg/day). The standard deviations for 20 to 200 kg of measurable tank inventory are shown in Figs. 1 and 2.

The data shows that for the low inventories (both tanks and contactors) the errors are throughput dominated for low and high throughput. At high throughput the throughput errors dominate at even high inventories (Fig. 2). For the low throughput case, tank inventory $\geq$40 kg, contribution from uncertainties in contactor inventory become significant ($\geq$0.5 kg) only for contactor inventories $\geq$10 kg, and then only for uncertainties $\geq$50%.

Similar analyses assuming uncertainties in contactor inventory of 25% rather than 10% are shown in Figs. 3 and 4. For the high throughput case (Fig. 4), uncertainties still are throughput dominated. For the low throughput case (Fig. 3), contactor inventory uncertainty can become significant ($\geq$0.5 kg) for the case of low tank inventory when contactor inventory exceeds ~5 kg.

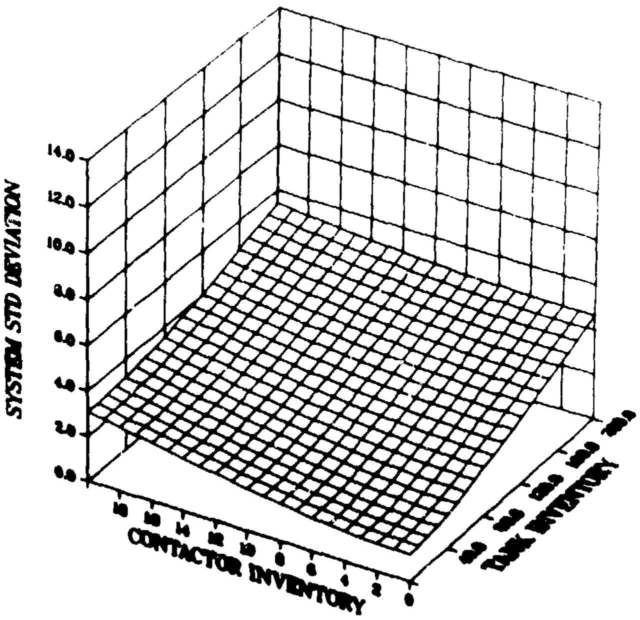

Fig. 1. Process standard deviation (kg) as a function of measurable tank inventory and unmeasurable contactor inventory, 5 kg/day throughput (150 kg/month); contactor inventory uncertainty is 10%.

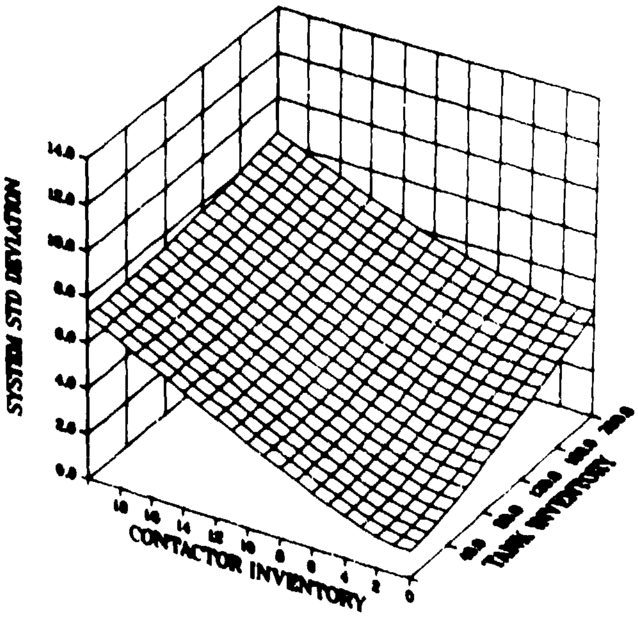

Fig. 2. Process standard deviation (kg) as a function of measurable tank inventory and unmeasurable contactor inventory, 50 kg/day throughput (1500 kg/month); contactor inventory uncertainty is 10%.

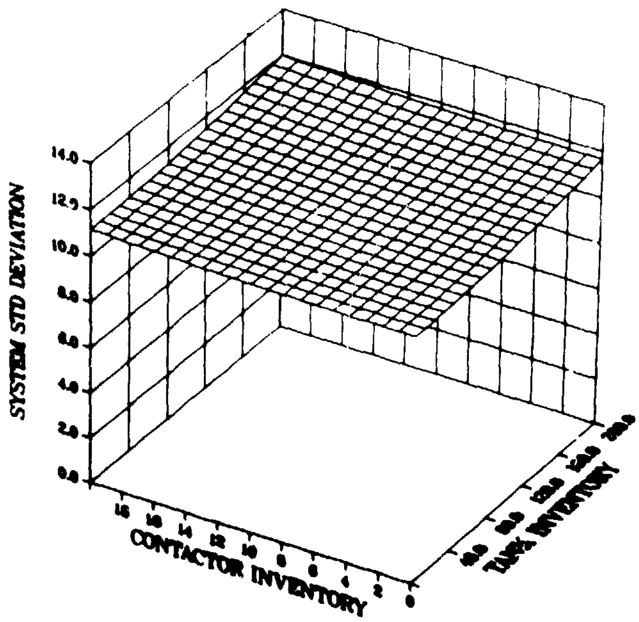

Fig. 3. Process standard deviation (kg) as a function of measurable tank inventory and unmeasurable contactor inventory, 5 kg/day throughput (150 kg/month); contactor inventory uncertainty is 25%.

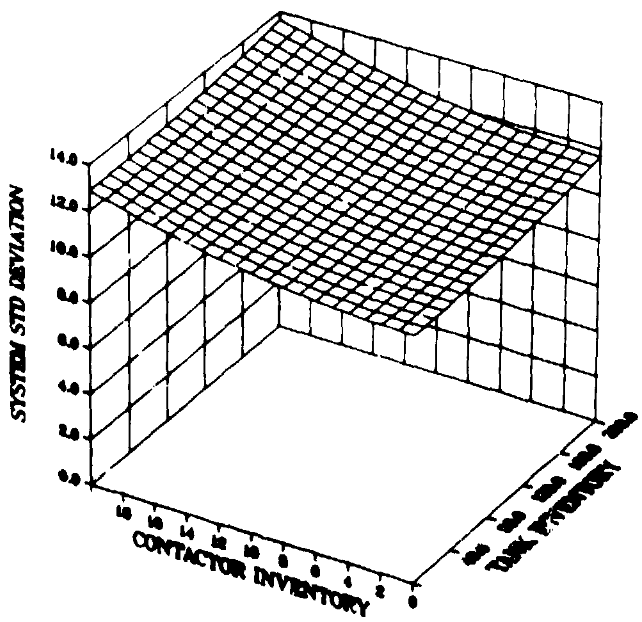

Fig. 4. Process standard deviation (kg) as a function of measurable tank inventory and unmeasurable contactor inventory, 50 kg/day throughput (1500 kg/month), contactor inventory uncertainty is 25%.

CONCLUSIONS

This type of approach can be used to assess the significance of contactor inventory uncertainty to overall system measurement uncertainties for any facility design. If the contribution from contactor inventory uncertainty is small relative to throughput and measured tanks, the need for independent inspector verification of these measurements becomes questionable. From verification of plant design, the inspector may be able to assume declared flow sheet values for contactor inventory. Application of this approach will be facility specific for any specific design, and the facility should be modeled before conclusions can be drawn on its applicability.

ACKNOWLEDGMENTS

The authors acknowledge the assistance of Lynne Williams for computer graphic support and Sharon Hurdle for manuscript preparation.

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